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ESRF

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Growth of Ge quantum dot lattices in alumina matrix	**Experiment number:** Si21-51
Beamline: BM20	**Date of experiment:** from: 27.10.2010 to:02.11.2010 from: 07.06.2011 to:14.06.2011	**Date of report:** 01.03.2012
Shifts: 18+18	**Local contact(s):** Carsten Baehtz	*Received at ESRF:*

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Report:

Materials consisting of regularly ordered quantum dots (QDs) in solid amorphous matrices attract great attention due to their interesting properties and many practical applications. The most attractive feature of such materials is possibility to design their properties by QD composition, size and arrangement. Another important key for design the material properties, is the matrix where QDs are buried. Ge is one of the most interesting semiconductor materials that can be used as building element for QDs due to very strong confinement effects and other interesting size-tuneable properties. Alumina is excellent choice for the matrix that hosts the QDs, because it has excellent mechanical and optical properties and it is also very compatible to Ge QDs for design of the electrical properties of the final material.

In our previous works we have analyzed the ordering and size properties of Ge QDs formed in a single continuous Ge+Al_2O_3 film [1] and in (Ge+Al_2O_3)/Al_2O_3 multilyer [2] deposited by magnetron sputtering deposition. We have demonstrated self-assembled growth in those systems, that was induced by a combination of diffusion mediated nucleation and influence of surface morphology on the nucleation positions.

In this work we have examined the growth and ordering properties of (Ge+Al_2O_3)/Al_2O_3 multilayer deposited also by magnetron sputtering deposition but with different geometry than the one used in the previous works. The main differences are: (i) the much closer distance between substrate and sputtering targets and (ii) the substrate stage which was kept fixed during the deposition. Additionaly, we made in-situ measurements that enabled us monitoring the changes in layer growth and its roughness properties. For the case of the multilayer investigated in Ref. [2], the substrate stage was rotating during the deposition and the distance target- substrate was much larger. We have shown that the deposition geometry influences significantly the ordering and shape properties of the QDs. We have explained the observed behavior by directional diffusion of Ge adatoms during the deposition.

The films consisting of twenty (Ge+Al_2O_3)/Al_2O_3 bi-layers with various layer thicknesses, were deposited by magnetron sputtering deposition at substrate temperature held at 300^{o}C. The deposition was performed at ESRF Grenoble, BM 20 beamline. During the deposition we have measured X-ray reflectivity (XRR) - time scans. For the deposition of Ge and Al_2O_3 we have used RF (3W) and (100W DC) magnetrons respectively. The Ar pressure was 5.4×10^{-3} mbar, and base pressure was 1×10^{-6} mbar.

The deposition process was in-situ monitored by XRR measurements. The incidence angle was kept fixed at 1 deg. This allows us to follow the evolution of multilayer growth including thicknesses of layers and surface roughness properties. The experimentally measured and simulated maps, showing the dependence of the measured XRR intensity on the deposition time for the multilayers, is shown in Fig. 1. The intensity profiles taken at α_f=0.25 and α_f=1 deg are depicted in Fig. 1(b). The oscillations are visible in both profiles, with the period equal to the period of magnetron shutters. Appearance of Bragg sheet caused by the multilayer growth and possible correlation of the QD positions is visible at α_f=0.8 deg.

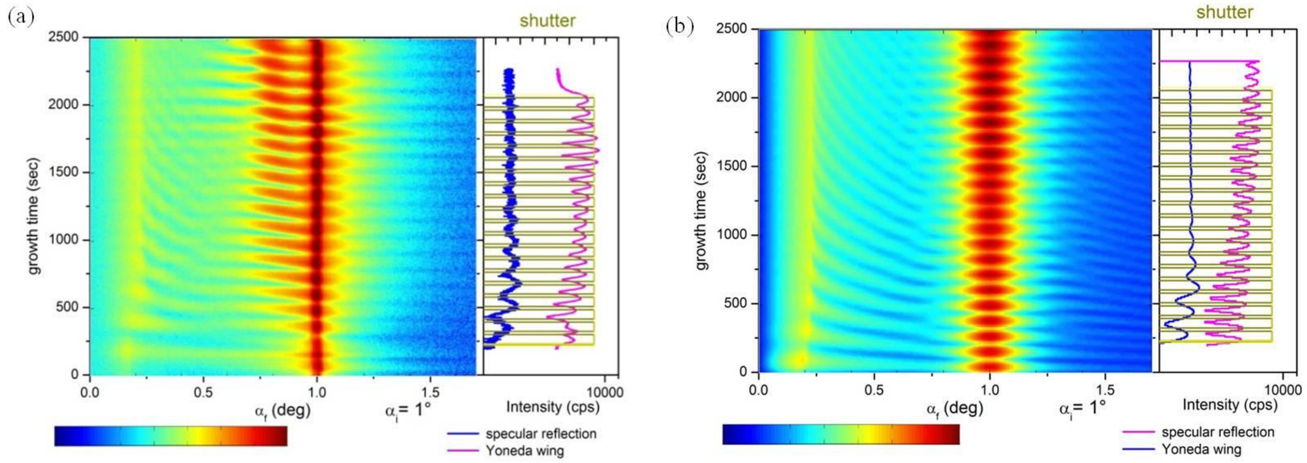

Figure 1. Experimentally measured time-scans of XRR on the growing films (multilayer left, a continous Ge+Al_2O_3 layer right) measured for incidence angle α_i=1 deg.

We have simulated the experimentally measured map using the standard theory for x-ray reflectivity; the simulation is shown in Fig. 1(b). The simulation corresponds well to the all important features of the experimentally measured map, except the Brag sheet at 0.8 deg. The reason is probably strong contribution of the correlation of the QDs positions in addition to the usual interface roughness features. The evolution of surface roughness during the growth process is found from the fit of the specular reflection. From the fit follows that periods of roughening and smoothing of the growing surface occurs during the deposition process. The surface roughness evolution is shown in Fig. 2. From the figure follows that surface roughness increases during the deposition of Ge-rich layer, while the deposition of pure Al_2O_3 layer causes smoothing of the growing surface. Such behavior can be related to self assembled growth of the QDs that is found for this film.

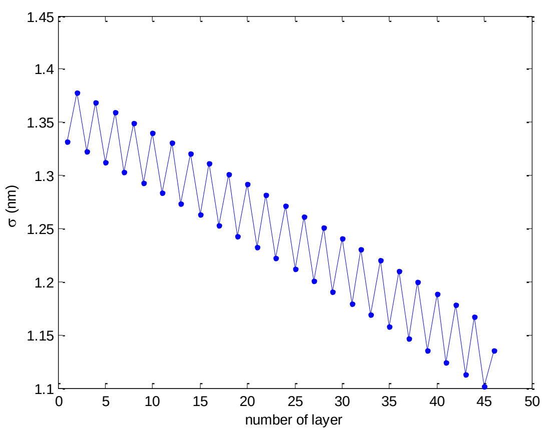

Figure 2. Dependence of interface roughness parameter σ on number of the deposited layer.

The analysis of the films structure after the deposition process was performed by grazing incidence small angle x-ray scattering (GISAXS) and transmission electron microscopy (TEM) techniques. From the analysis follows formation of a three-dimensional crystal-like lattice of quantum dots, with certain degree of anisotropy caused by the specific deposition conditions. The observed regular ordering follows from the interplay of diffusion mediated nucleation and surface morphology features that are caused by the formation of Ge clusters. Thus, the surface roughness increases during the growth of Ge-rich layer. The anisotropy is explained by a directional diffusion of Ge adatoms during the deposition process, caused by preferential direction of Ge atoms from the magnetron sputtering target [3].

[1] Buljan, M.; Pinto, S. R. C.; Rolo, A. G.; Martin Sanchez, J.; Gomes, M. J. M.; Mücklich, A.; Bernstorff, S.; Holy, V. *Phys. Rev. B* **2010**, *82*, 235407.
[2] M. Buljan et al. Germanium Quantum Dots in Alumina-Based Multilayers: Self-Assembly for Advanced Properties, submitted, (2012).
[3] M. Buljan, C. Baehtz, et al. Growth of Ge quantum lattice in an amorphous alumina matrix investigated by in-situ x-ray scattering. In preparation.